

# High-pressure phase relations of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ single crystals

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We have investigated the pressure dependence of the superconducting transition temperature  $T_c$  up to 18 GPa of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals ranging from the highly underdoped through the nearly optimally doped to the highly overdoped level. For all three samples studied,  $T_c$  is found to increase initially and then saturate at a critical pressure  $P_c$  but decrease modestly with further compression. Oxygen doping tends to reduce the increase in  $T_c$  and  $P_c$ . A new high-pressure phase diagram between the saturated  $T_c$  and  $P_c$  is then obtained. Theoretical interpretation is given by using the competition between the hole carrier density and pairing interaction strength based on the high-pressure transport data of the resistivity and Hall coefficient in this system.

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## I. INTRODUCTION

The doping dependence of the high-temperature superconductors (HTSCs) has become a significant issue due to pseudogap phenomena in underdoped compounds and to the peculiar influence of hole carrier density on properties of superconducting and normal states. Among the HTSCs discovered so far,  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ ,  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  are the mostly extensively studied systems due to available single crystals over a large range of doping levels and transition temperatures.  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  has a structural phase transformation between the underdoped and overdoped regimes.<sup>1</sup> Meanwhile, changing carrier density through cation substitution probably leads to local distortion due to the size effect.<sup>2</sup> Although the carrier density in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  can be tuned by oxygen content, the occurrence of chain oxygen sites in this material complicates interpretation of much of the experimental data. Moreover, it is difficult to make even slightly overdoped samples in this system, only the underdoped part of the phase diagram can be thoroughly studied. By comparison,  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  is a perfect candidate for studying its physical properties over a wide range of the doping.

In order to get insight into the basic mechanism responsible for high-temperature superconductivity, one always strives to observe the possible change of the superconducting transition temperature  $T_c$  at fixed doping level. Pressure has been realized to be an effective way to change  $T_c$  for a compound. However, the effect of pressure on  $T_c$  is very complicated. The pressure derivative of  $T_c$ ,  $dT_c/dP$ , can vary from positive to negative, which strongly depends on the doping level. For most optimally doped compounds,<sup>3,4,5,6,7,8,9</sup>  $T_c$  generally increases when pressure is applied at the initial stage, passes through a saturation at some critical pressure level  $P_c$ , then decreases modestly at high pressures.  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  is the only known system where the constantly positive  $dT_c/dP$  is not sensitive to oxygen content,<sup>10,11,12</sup> which

makes it an ideal system to investigate the phase diagram between the saturated  $T_c$  and critical pressure  $P_c$  over an entire doping regime. Early measurements on the 80~88 K samples<sup>13,14</sup> have shown that for the sample with a relatively high  $T_c$  it is easy to reach the saturation at the beginning of the pressure range. It was also shown that the  $P_c$  shifts to higher pressure with additional oxygen content in the overdoped side.<sup>13,14</sup> It remains unclear whether the saturation in the  $T_c$  versus  $P$  curve is present for samples away from optimal doping. Therefore, it is of interest to study the pressure dependence of  $T_c$  as well as the high-pressure phase diagram in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  over the entire doping range.

Experiments carried out on a variety of HTSCs<sup>5,15</sup> have revealed that  $T_c$  is not a unique function of pressure but depends strongly on the temperature at which the pressure is applied. For almost all systems, pressure-induced relaxation effects become activated for temperatures above 200 K. The pressure derivative  $dT_c/dP$  can be increased tenfold if the pressure is changed at room temperature rather than at low temperatures.<sup>16</sup> Since the relaxation is believed to be frozen at low temperatures, the pressure dependence of  $T_c$  and the pressure derivative  $dT_c/dP$  basically represent the intrinsic pressure effect. Such an effect most directly reflects changes in the basic physical properties, including the pairing interaction itself, and thus is most valuable for comparison with theory. It is therefore of interest to perform detailed measurements of  $T_c(P)$  under high pressure by changing pressure at low temperatures to elucidate the intrinsic pressure effects.

In this paper we present the results of a high-pressure study of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals in which the pressure is applied and measured at low temperatures. The characteristic nonmonotonic pressure dependence of the superconducting transition temperature is found in our crystals ranging from heavily underdoped to heavily overdoped samples. We obtain a new high-pressure phase diagram between the saturation of  $T_c$  and critical pres-

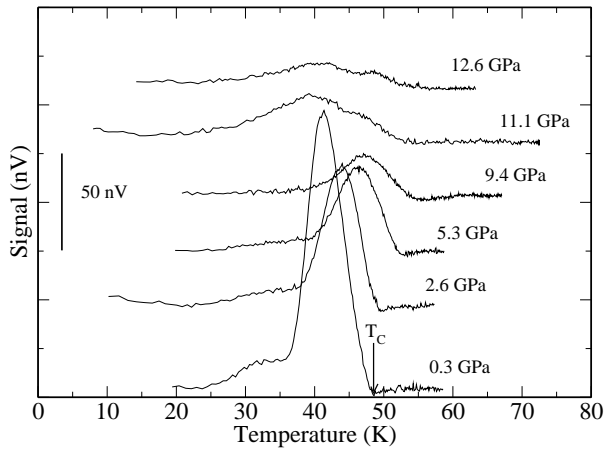


FIG. 1: Magnetic susceptibility signal in nV vs. temperature for an underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  crystal at various pressures.

sure  $P_c$  at which  $T_c$  is saturated. We demonstrate that the nonmonotonic behavior originates from a competition between the hole carrier density and pairing interaction strength.

## II. EXPERIMENTAL

Samples of single crystal  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  were grown by a self-flux technique in a strong thermal gradient to stabilize the direction of solidification using a stoichiometric ratio (Bi:Sr:Ca:Cu=2:2:1:2) of cations, as described elsewhere.<sup>17</sup> Vacuum anneals are carried out in sealed quartz tubes to obtain the underdoped crystals. Overdoping has been accomplished using stainless steel cells sealed with samples immersed in liquid oxygen. Crystals used in the high-pressure studies are the highly underdoped (“vacuum-annealed,”  $T_c = 48$  K), air-annealed ( $T_c = 86$  K) and overdoped (“oxygen-annealed,”  $T_c = 60$  K) samples. A 10-90% transition width of 0.5 K in susceptibility of our samples is indicative of very good quality and of high homogeneity in both cation and oxygen concentrations.<sup>17</sup>

We performed the measurements of  $T_c$  under high pressures using a highly sensitive magnetic susceptibility technique with diamond anvil cells.<sup>18</sup> This technique has been proven to be especially successful in the discovery of superconductivity in compressed sulphur and lithium.<sup>19</sup> The technique is based on the quenching of the superconductivity and suppression of the Meissner effect in the superconducting sample by an external magnetic field. The magnetic susceptibility of the metallic parts of the high-pressure cells is essentially independent of the external field. Therefore, the magnetic field applied to the sample inserted in the diamond cell mainly affects the change of the signal coming from the sample. If we apply an alternating low-frequency ( $f = 20$  Hz) magnetic

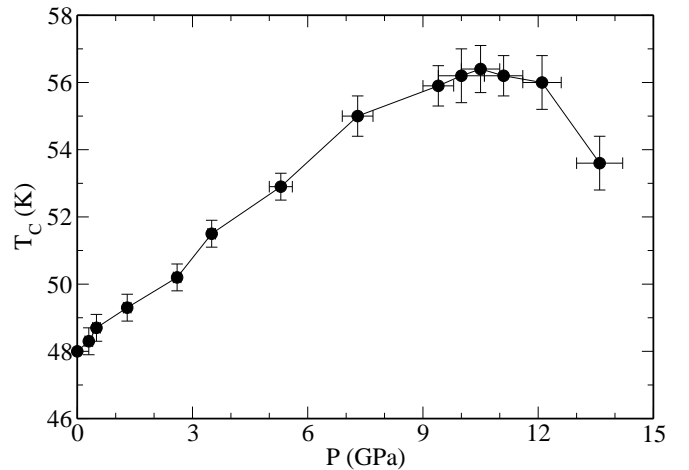


FIG. 2: Dependence of the superconducting transition temperature  $T_c$  on pressure in an underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal.

field with an amplitude below 100 Oe, then the output signal changes twice per period of the magnetic field, because the positive and negative magnetic fields act identically to destroy the superconducting state. The resulting high-frequency output signal is modulated in amplitude at frequency  $2f$ . This modulation is observed only in the narrow range of temperatures close to the temperature of superconducting transition. The  $T_c$  is then identified as the point where the signal goes to zero because of the disappearance of the Meissner effect.

Samples are loaded in Mao-Bell cells made from hardened Be-Cu alloy. The gaskets are made of nonmagnetic Ni-Cr alloy. The  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  crystal (typical size  $50 \times 50 \times 10 \mu\text{m}^3$ ) together with ruby chips are placed in the sample chamber. NaCl is loaded into the gasket hole with diameter  $200 \mu\text{m}$  to serve as pressure medium. The pressure is applied and measured at low temperatures and is gauged by the  $R1$  fluorescence line of ruby.<sup>20</sup> Each initial pressure run was performed as single run at low temperature, pressure was changed immediately after the temperature scan was finished, and sample was taken back to low temperature to start another temperature scan.

## III. RESULTS

Figure 1 shows the representative temperature scans at different applied pressures for an underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal. Superconducting transition is identified as the temperature where the signal goes to zero on the high-temperature side which is the point at which magnetic flux completely enters the sample. The superconducting transition of 48 K is obtained at ambient pressure. It is clear that at the pressure of 9.4 GPa the superconducting transition shifts to higher tem-

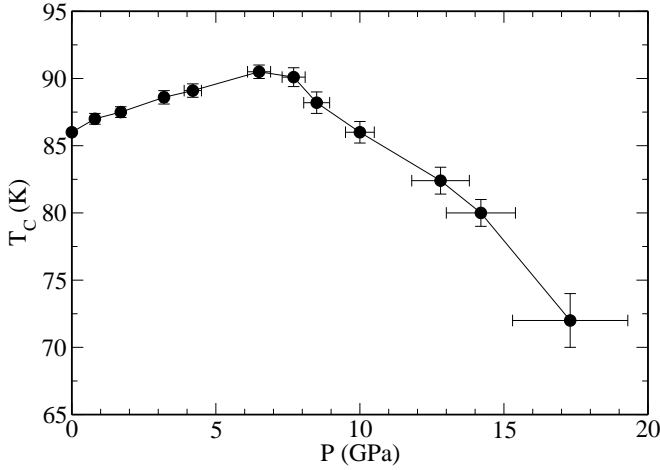


FIG. 3: Shift of the superconducting transition temperature  $T_c$  with applied pressure up to 18 GPa for a  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal which is nearly optimally doped but slightly overdoped.

peratures than at 1 atm, but it turns back beyond that pressure. The signal amplitude is suppressed by further increasing pressure. Also, at high pressures the shape of the signal broadens. In our measurements, we apply pressure in the increasing run and at low temperatures below 100 K. Therefore, we are confident for the existence of pressure medium over the whole measured range.

In Fig. 2, we plot the obtained  $T_c$  values in this underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  sample as a function of pressure. As shown,  $T_c$  is initially increased with applied pressure passing through a saturation value at around  $P_c = 10.5$  GPa and then decreases at higher pressures. A positive initial pressure derivative of 1.5 K/GPa is obviously consistent with the pure hydrostatic pressure measurements with the helium gas system.<sup>10</sup> The nonmonotonic pressure dependence of  $T_c$  observed in the present heavily underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  is believed to be a common characteristic for all underdoped  $p$ -type copper-oxides. This behavior has also been reported in the  $\sim 80$  K  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  samples with nearly optimal dopings.<sup>13,14</sup> For an underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal with  $T_c = 54$  K which is close to our sample, Klotz and Schilling<sup>14</sup> found an initial constancy of  $T_c$  up to 1.6 GPa as well as the absence of the saturation up to 6 GPa. This difference might be due to the difference of the samples and/or the difference in the hydrostaticity of the pressure medium used.

The magnetic susceptibility curves for a nearly optimally doped but slightly overdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal have a behavior similar to that in the underdoped sample. The superconducting transition is at 86 K at ambient pressure and shifts upward when pressure is applied at the beginning stage. Above 8.5 GPa, further pressure moves the transition to lower  $T_c$  values. The amplitude of the signal becomes weak with the ap-

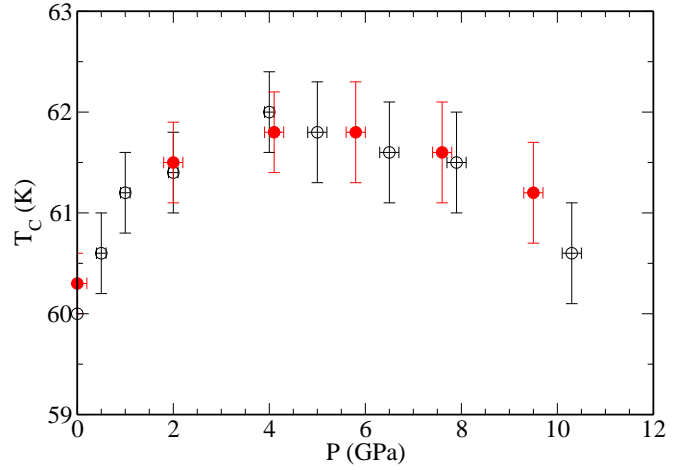


FIG. 4: (Color online) Superconducting transition temperature  $T_c$  for an overdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  sample as a function of pressure. The open and solid circles represent the measurements in the compressing and decompressing run, respectively.

plied pressure. It is hard to distinguish the signals from the sample or background at pressures above 18 GPa.

Shown in Fig. 3, the pressure dependence of  $T_c$  displays the characteristic behavior as is in other optimally doped HTSCs.<sup>3,4,5,6,7,8</sup> Similar nonmonotonic pressure dependence of  $T_c(P)$  with a saturation at a critical pressure near 2 GPa was reported in the sister bilayer  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_{8+\delta}$  system at the optimal doping.<sup>3</sup> For the initial slope of the  $T_c$  versus  $P$  curve in Fig. 3 we derive  $dT_c/dP = 1.3$  K/GPa similar to that measured in the helium gas system.<sup>10</sup> The saturation occurs at  $P_c \approx 6.5$  GPa in our  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  sample at near optimal doping. This critical value is approximately the same as that in the  $T_c \sim 80$  K crystal of Alekseeva et al.,<sup>13</sup> but relatively higher than that for the  $T_c = 88$  K sample of Klotz and Schilling.<sup>14</sup>

We have measured the temperature dependence of magnetic susceptibilities near  $T_c$  for an overdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal in both the compressing and decompressing runs. As plotted in Fig. 4, the susceptibility curves are nearly identical, regardless of the pressure history. In the decompressing run,  $T_c$  almost returns to its original ambient pressure value upon full release of the pressure. This is indicative of free relaxation in this sample.

From Fig. 4, it is found that as the applied pressure is increased,  $T_c$  continues to increase until it reaches a saturation value at near 4 GPa. Previous studies<sup>14</sup> on an overdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal with a relatively high  $T_c = 80$  K have shown that the  $T_c$  also has a saturation value at near  $P_c = 4$  GPa. Compared to the maximum value of  $T_c \approx 90$  K in flux-grown  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  at optimal doping, the present sample with a  $T_c$  of 60 K is heavily overdoped. The occur-

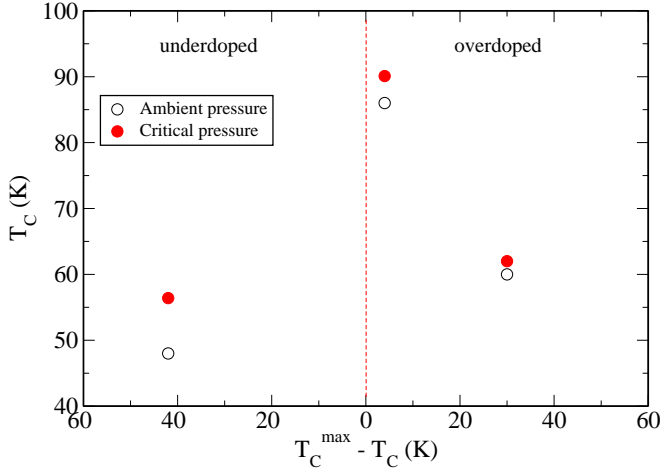


FIG. 5: (Color online) Plot of the superconducting transition temperature  $T_c$  at the ambient pressure (open circles) and critical pressure  $P_c$  (solid circles) in flux-grown  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals as a function of  $T_c$  referenced to the maximum value of 90 K.

rence of the saturation in the  $T_c(P)$  curve in our sample is very interesting, since it has never been reported in such a heavily overdoped cuprate where the significant change of  $T_c$  is tuned only through oxygen content rather than cation substitution. Considering the fact that  $T_c$  would decrease with further adding oxygen content in the overdoped regime, this result clearly indicates that neither the pressure-induced charge transfer<sup>21</sup> nor the pressure-induced oxygen doping<sup>22</sup> is solely responsible for the pressure effect on  $T_c$ . The observed saturation in the  $T_c(P)$  curve also suggests that another intrinsic variable independent of the hole carrier density is playing a significant role in enhancing  $T_c$  at even the heavily overdoped region.

Among the results obtained in these experiments, the most important finding is that the saturation with larger value than the ambient  $T_c$  occurs in the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  samples over the entire doping range from the heavily underdoped through the optimally doped to the heavily overdoped. Both the critical pressure  $P_c$  and difference between the  $T_c$  and its saturated value decrease with the hole carrier density created by adding oxygen into the  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  system. This behavior is similar to that reported for the  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  system in the overdoped side<sup>23</sup> where the pressure-induced transition between the orthorhombic and tetragonal phases is accompanied when  $T_c$  reaches saturation.<sup>24</sup> Furthermore, we find the similar trend of  $T_c(P_c)$  in the present  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  samples at different doping levels as observed in other optimally doped compounds.<sup>3,4,5,6,7,8,9</sup> That is, the higher the  $P_c$ , the larger the saturation value of the  $T_c$  is. We thus obtain an extended phase diagram at both the ambient condition and critical pressure, which is presented in Fig. 5. The saturation of  $T_c$  under the critical pressure of a hole-

doped cuprate has been shown to cover the heavily underdoped to heavily overdoped regime. Despite extensive effort to reaching the saturation at the critical pressure, such a high-pressure phase diagram over the whole doping regime has not been established previously. The saturation  $T_c(P_c)$  was only found in either the underdoped side<sup>5</sup> or the overdoped side.<sup>14,23</sup>

#### IV. DISCUSSION

To understand the experimentally observed nonmonotonic behavior of  $T_c(P)$  of these samples, we use a simple  $d$ -wave BCS formalism:

$$1 = \frac{1}{2N} \sum_k \frac{Vg^2(k)}{|\varepsilon_k - \mu|} \tanh\left(\frac{|\varepsilon_k - \mu|}{2k_B T_c}\right). \quad (1)$$

Where  $g(k) = \cos k_x - \cos k_y$ ,  $\varepsilon_k$  is the quasiparticle dispersion,  $N$  is the number of  $k$  vectors,  $k_B$  is the Boltzmann constant, and  $V$  is the in-plane pairing interaction strength. The constraint condition for the hole carrier density  $n_H$  in conjunction with the chemical potential  $\mu$  is given by

$$n_H = \frac{1}{2} - \frac{1}{2N} \sum_k \tanh\left(\frac{|\varepsilon_k - \mu|}{2k_B T_c}\right). \quad (2)$$

The validity of this kind of  $d$ -wave BCS formalism in describing the superconducting state of HTSCs has been confirmed by recent angle-resolved photoemission spectroscopy (ARPES)<sup>25</sup> and transport<sup>26,27</sup> measurements. As quasiparticle dispersion, we use<sup>28</sup>  $\varepsilon_k = 0.166 \cos k_x \cos k_y + 0.046(\cos 2k_x + \cos 2k_y)$  eV, which corresponds to the hole dispersion in an antiferromagnetic background at half-filling, but should be approximately valid also at finite  $n_H$  as long as the antiferromagnetic correlation length is not negligible. A characteristic feature of this dispersion is the near flatness of the energy in the vicinity of  $(\pi, 0)$ , which is in good agreement with ARPES results for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ .

The experimentally observed parabolic relation between  $T_c$  and  $n_H$  can be obtained by combining Eqs. (1) and (2) for a given  $V$ . Taking the maximum  $T_c$  of 90 K for flux-grown  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ , we deduce  $V = 0.03782$  eV. The resulting carrier densities  $n_H$ 's are then determined for the samples with various  $T_c$  values.

The temperature dependences of both the in-plane resistivity  $\rho_{ab}$  and Hall coefficient  $R_H$  under high pressures have been suspected of giving clues to the pressure effects on  $T_c$ . For optimally doped samples at ambient pressure, the basic experimental trends are  $\rho_{ab} \propto T$  and  $R_H \propto 1/T$  in the high temperature regime around room temperature. The quantity  $R_H e z / v$  is inversely proportional to the hole carrier density  $n_H$ , where  $z$  is the number of Cu atoms per unit cell of volume  $v$  and  $e$  the carrier charge. In the high-temperature  $T$ -linear region, the resistivity satisfies the simple form  $\rho_{ab} = (4\pi/\omega_p^2)\tau^{-1}$ ,

where  $\omega_p$  is the plasma frequency and the scattering rate  $\tau^{-1} = 2\pi\Lambda T$  with  $\Lambda$  being the coupling strength between the charge carriers and some excitations. We take the Drude spectral weight  $\omega_p^2 \sim n_H/m^*$  with  $m^*$  being the carrier effective mass. Systematic resistivity studies suggest that both the carrier scattering rate and coupling strength are independent of doping and nearly same for a superconductor.<sup>29</sup> The pairing interaction strength  $V$  appeared in Eq. (1) should have the same physical meaning as  $\Lambda$ . Therefore, we have  $\rho_{ab} \propto (m^*/n_H)VT$  and  $R_H \propto v/(n_H ez)$ , which should account for the resistivity and Hall data at room temperature and under high pressures. Taking the pressure derivative of the quantities in the expressions of  $\rho_{ab}$  and  $R_H$ , we have the following relations:

$$\frac{d \ln \rho_{ab}}{dP} = \frac{d \ln m^*}{dP} - \frac{d \ln n_H}{dP} + \frac{d \ln V}{dP}, \quad (3)$$

$$\frac{d \ln R_H}{dP} = -\kappa_v - \frac{d \ln n_H}{dP}. \quad (4)$$

Where the volume compressibility  $\kappa_v \equiv 1/B_0 = -d \ln v/dP$  and  $B_0$  is the bulk modulus.

For the present  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  system, we take  $d \ln R_H/dP = -10.8 \times 10^{-2} \text{ GPa}^{-1}$  and  $\kappa_v = 16.7 \times 10^{-3} \text{ GPa}^{-1}$  from the Hall coefficient<sup>30</sup> and neutron diffraction<sup>31</sup> measurements, respectively. Substituting these values into Eq. (4), we obtain  $d \ln n_H/dP = 9.1 \times 10^{-2} \text{ GPa}^{-1}$ . Studies of pressure effects on the irreversibility line<sup>32</sup> in a nearly optimally doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  reveal that the in-plane penetration depth  $\lambda_{ab}$  decreases from 220 to 205 nm when the applied pressure increases from 0 to 1.5 GPa, yielding  $d \ln \lambda_{ab}/dP = -4.55 \times 10^{-2} \text{ GPa}^{-1}$ . As it is known  $\lambda_{ab}$  can be expressed in the following relation:  $\lambda_{ab}^2 = m^*/n_H$ . We then get  $d \ln m^*/dP = 2d \ln \lambda_{ab}/dP + d \ln n_H/dP$ . According to this simple relation, we obtain the value of  $d \ln m^*/dP$  of zero by using the experimental data for  $d \ln \lambda_{ab}/dP$  and  $d \ln n_H/dP$ . It is therefore reasonable to neglect the pressure dependence of  $m^*$ . This indicates that the hole carrier density  $n_H$  and pairing interaction strength  $V$  are the two main intrinsic pressure variables responsible for pressure effect besides the cell volume  $v$ . This conclusion drawn from the normal-state transport properties agrees very well with the assumption in interpreting the pressure effect in the Y-Ba-Cu-O systems.<sup>33</sup>

High-pressure transport measurements give  $d \ln \rho_{ab}/dP = -7.5 \times 10^{-2} \text{ GPa}^{-1}$  for a nearly optimally doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystal at 300 K.<sup>12</sup> Then we have  $d \ln V/dP = 1.6 \times 10^{-2} \text{ GPa}^{-1}$  by using Eq. (3). This result implies  $d \ln V/dP \simeq \kappa_v$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ . Noting that the pressure-induced relative change of  $V$  satisfies a simple formalism  $d \ln V/dP = (2 \sim 4)\kappa_a$  in the Y-Ba-Cu-O systems,<sup>33</sup> where  $\kappa_a \equiv -d \ln a/dP$  being the lattice compressibility along the  $a$  axis. The relationship between the pairing interaction strength and cell volume is then expressed by  $\partial \ln V/\partial \ln v \simeq -1$ . We would like to mention that this probably universal relation does not rely on any model,

at least in these two most generally studied HTSCs.

Since  $n_H$  and  $V$  are two intrinsic pressure variables, the variation of  $T_c$  with pressure is believed to be due to the change of the competition between them. Assuming that the pressure dependence of both  $n_H$  and  $V$  are not as linear in pressure  $P$ , but in the relative volume decrease given by  $1 - v(P)/v_0$ , we can write  $n_H(P)$  and  $V(P)$  as

$$\begin{aligned} n_H(P) &= n_H(0) \left[ 1 + \frac{d \ln n_H}{dP} B_0 \left( 1 - \frac{v(P)}{v_0} \right) \right], \\ V(P) &= V(0) \left[ 1 + \frac{d \ln V}{dP} B_0 \left( 1 - \frac{v(P)}{v_0} \right) \right]. \end{aligned} \quad (5)$$

The pressure dependence of the relative volume can be well described by the first-order Murnaghan equation of state  $v(P)/v_0 = (1 + B'_0 P/B_0)^{-1/B'_0}$ , where  $B'_0$  is the pressure derivative of  $B_0$  and  $v_0$  the cell volume at ambient pressure. It has been shown that the construction of the pressure dependence of the intrinsic pressure variable through the relative volume change always reproduces an excellent agreement of the resulting curve with the experimental values.<sup>34,35</sup> Olsen *et al.*<sup>36</sup> obtained  $B'_0 = 6.0$  for Bi-based cuprates by fitting their high-pressure X-ray diffraction data up to 50 GPa. Their bulk modulus  $B_0$  coincides well with that derived from the neutron diffraction technique.<sup>31</sup>

The competition between the  $n_H(P)$  and  $V(P)$  provides natural explanation for the observed high-pressure phase diagram as shown in Fig. 5.  $V(P)$  is a favorable factor for the increase of  $T_c$  with pressure. While,  $n_H(P)$  favors  $T_c$  only for the compound in the underdoped regime, and it becomes a negative factor in the optimally doped or the overdoped region. For the underdoped compound,  $n_H(P)$  would be the negative factor once the pressure-induced increase of  $n_H$  passes through the optimal doping. The crucial parameter  $P_c$  is the crossover pressure where the factor(s) increasing  $T_c$  is(are) equal to that(those) decreasing  $T_c$ . A higher  $P_c$  in our heavily underdoped sample arises from both the positive contribution to  $T_c$  from  $V(P)$  as well as the relatively long way for  $n_H$  to go before passing through the optimal doping. The nearly optimally doped compound has a modest  $P_c$  due to the favorable factor  $V(P)$  but the unfavorable  $n_H(P)$ . Although  $V(P)$  still factors the  $T_c$  increase for the heavily overdoped material, the pressure-induced increase of  $n_H$  strongly suppresses such  $T_c$  increase. As a result, there only exists a smaller  $P_c$  for the heavily overdoped material. Therefore, the extended high-pressure phase diagram can be well understood on the basis of the competition of the two intrinsic pressure variables.

Using the determined parameters  $B_0$ ,  $B'_0$ ,  $d \ln n_H/dP$ , and  $d \ln V/dP$ , we are able to evaluate the pressure dependence of  $T_c$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  samples in terms of Eqs. (1), (2), and (5). In Fig. 6 we plot the calculated variation of  $T_c$  as a function of pressure up to 20 GPa for the optimally doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  compound with  $T_c = 90$  K at ambient condition. As can be seen, as pressure is increased,  $T_c$  increases initially until

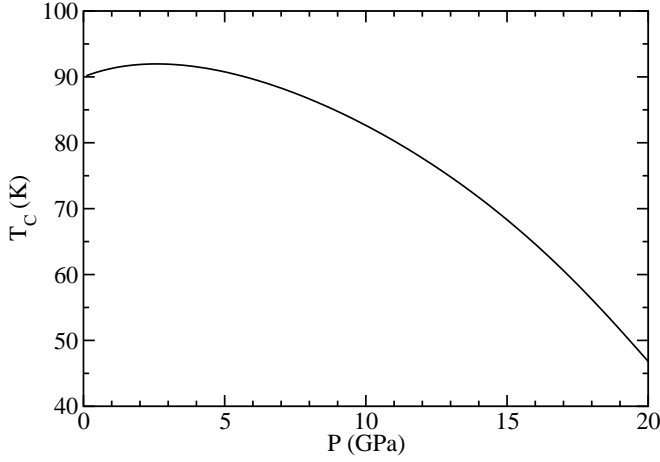


FIG. 6: Calculated variation of the superconducting transition temperature  $T_c$  with pressure for the optimally doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ .

passing a saturation at critical pressure  $P_c$  of 2.6 GPa, and at higher pressures  $T_c$  decreases. This nonmonotonic behavior of  $T_c(P)$  of the optimally doped compound is consistent with the measurements in our nearly optimally doped sample as well as the 88 K sample of Klotz and Schilling.<sup>14</sup> The theoretical value of the initial  $dT_c/dP$  is 1.69 K/GPa for the optimally doped material, which is in excellent agreement with the reported  $1.5 \pm 0.2$  K/GPa from various measurements.<sup>10,11,12</sup> It is apparent that the competition between the hole carrier density and pairing interaction strength indeed captures the essential physics of pressure effect on  $T_c$ .

Figure 7 shows the critical pressure  $P_c$  dependence of the difference between the superconducting transition temperatures  $\Delta T_c$  at the ambient pressure and critical pressure in the optimally doped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  with those in some other optimally doped compounds.<sup>3,4,5,6,7,8</sup> It can be seen that there exists the same trend of increasing the  $\Delta T_c$  value with the increase of  $P_c$  in these optimally doped compounds.  $\Delta T_c$  increases under  $P_c$  at the rate of near 1 K/GPa. It is indicated that any optimally doped compound has a saturation value of the maximum  $T_c$  which can be reached by applied pressure at critical  $P_c$  value. Moreover, it is confirmed that pressure is a most effective way to enhance  $T_c$  in optimally hole-doped cuprates. These results also suggest that the enhancement of the maximum  $T_c$  with pressure in the optimally doped HTSCs is not due to the increase of the pressure-induced hole carrier density.

Since the above determined parameters are available only for the optimally or nearly optimally doped materials, we can not calculate the change of  $T_c$  with pressure for various dopings by using the same parameters, especially for  $d \ln n_H/dP$  and  $d \ln V/dP$ . There are not enough high-pressure transport data available in obtaining the information on these parameters at the moment. However, we can estimate the pressure-induced change

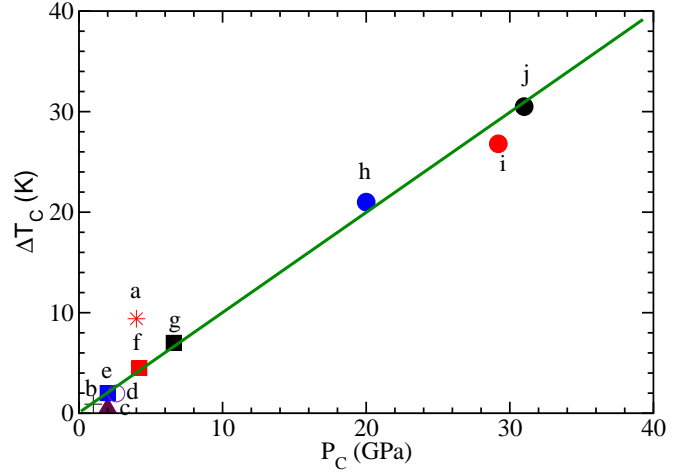


FIG. 7: (Color online) Critical pressure  $P_c$  dependence of the difference between the superconducting transition temperatures ( $\Delta T_c$ ) at the ambient pressure and critical pressure of optimally hole-doped HTSCs: a,  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (star, Ref. [4]); b,  $\text{La}_{2-x}\text{Ca}_{1+x}\text{Cu}_2\text{O}_6$  (plus, Ref. [6]); c,  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (triangle, Ref. [5]); d,  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (open circle, this work); e,  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Ref. [3]); f,  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$  (Ref. [7]); g,  $\text{Tl}_2\text{Ba}_2\text{Ca}_3\text{Cu}_4\text{O}_{12+\delta}$  (Ref. [7]); h,  $\text{HgBa}_2\text{CuO}_{4+\delta}$  (Ref. [8]); i,  $\text{HgBa}_2\text{CaCu}_2\text{O}_{6+\delta}$  (Ref. [8]); j,  $\text{HgBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$  (Ref. [8]). The line is the guide to the eyes.

of carrier density  $n_H$ , if we assume that  $d \ln V/dP$  is independent of oxygen content just as the interaction strength  $V$  is. Substituting the experimentally determined  $dT_c/dP$  of 1.5 K/GPa for  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ,<sup>10</sup> we get the  $d \ln n_H/dP$  values of  $5.1 \times 10^{-3}$ ,  $6.4 \times 10^{-3}$ , and  $2.5 \times 10^{-3} \text{ GPa}^{-1}$  for the underdoped, nearly optimally doped, and overdoped samples with  $T_c = 48$ , 86, and 60 K, respectively. For comparison, the high-pressure transport data provide the  $d \ln n_H/dP$  value of  $9.1 \times 10^{-2} \text{ GPa}^{-1}$  for the optimally doped compound with  $T_c = 90$  K. This means that the optimally doped compound has the largest amount of the pressured-induced increase of the hole carrier density among the samples with different doping level. However, as discussed above, the pressure effect on  $T_c$  in this material is still dominated by the pairing interaction strength.

## V. CONCLUSION

We have measured the pressure dependence of the superconducting transition temperature  $T_c$  in flux-grown  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  single crystals by using a highly sensitive magnetic susceptibility technique with diamond anvil cells. In order to avoid the possible contribution from the pressure relaxation effect, we apply and measure pressure at low temperatures. The nonmonotonic pressure behavior of  $T_c$  is observed for all the samples, from the heavily underdoped through the nearly opti-

mally doped to the heavily overdoped regime. We therefore obtain a high-pressure phase diagram between the saturation value of  $T_c$  and critical pressure  $P_c$ . Theoretical calculation based on a  $d$ -wave BCS formalism reproduces well this nonmonotonic behavior in terms of the high-pressure transport data. We have demonstrated that the pressure dependence of  $T_c$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  is the result of the competition between the hole carrier density and pairing interaction strength. The generally observed  $T_c(P_c)$  in our samples indicates that the pairing interaction strength is mainly responsible for the pressure effect in this system.

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